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Materials Structure Physics Research Group
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Research Activities

Preface

The chief aim in our researches is to construct "Orbital Physics" using synchrotron x-ray and neutron scattering techniques. This new physics is rapidly developing now. The orbital as well as charge and spin is a fundamental degree of freedom of an electron. The electronic orbital controlling the electron transfer has a great influence on the electronic and magnetic properties of the system. However, it has not been so easy task to detect the ordering of this degree of freedom (orbital ordering). We have experimentally developed some new techniques of synchrotron x-ray and neutron scattering. One of them is the resonant x-ray scattering (RXS) to detect the anisotropy of the scattering factor due to the orbital ordering. The orbital states of many compounds has been elucidated by these techniques so far.

This year some subjects of transition metal oxides have been continuously studied: In manganites new modulated structures were discovered and in titanates the orbital states near the metal-insulator transition were investigated by RXS. The films of manganites as well as bulk compounds were also examined and made the electronic state of the interface clear by using RXS. Some f-electron systems, RB_2C_2 and TmTe and so on, have been continuously studied by x-ray and neutron scattering. This year we expanded the subject of our study to a new f-electron system, Skutterudite, which is mainly studied by Iwasa's group.

We are collaborating with JAERI (Japan Atomic Energy Research Institute) group at SPring-8 on synchrotron x-ray experiments because Youichi MURAKAMI is a guest group leader of SRRC (Synchrotron Radiation Research Center) of JAERI. We have constructed a new beamline, BL22XU, and are carrying out the experiments on RXS with a low energy down to 3keV and resonant inelastic x-ray scattering. The high brilliance of this station also make it possible to measure the ordering under extreme conditions, such as high magnetic field, high pressure in low temperature.

This group has been reconstructed from April 2001 when Youichi MURAKAMI has come as a new group leader: Dr. Kazuma HIROTA and Dr. Masayasu TAKEDA moved into Institute of Solid State Physics, the University of Tokyo and JAERI, respectively, Dr. Hironori NAKAO transfered from Institute of Materials Structure Science, KEK. Finally, Dr. Kazuaki IWASA joined us from Tokyo Metropolitan University as a new Associate Professor in July, 2003. The reconstruction of this group has been completed.

Scientific activities

Charge and Orbital Ordering in $Nd_{1-x}Sr_{1+x}MnO_4$

(T. Sato, H. Nakao, Y. Murakami, H. Nakao)

We have studied the charge and orbital ordering in $Nd_{1-x}Sr_{1+x}MnO_4$ (x = 0.67, 0.75). The charge and orbital ordering of La1-xSr1+xMnO4 has been intensively investigated. However, the ordered states with x ¿ 0.5 have not been studied owing to the chemical phase separation. Recently the sample has been prepared by substituting Nd for La. The important question is whether the charge state (the ordering of Mn^{3+} and Mn^{4+}) of this sample is localized or delocalized one. The orbital state is also interesting concerned with the type of the ordering, that is, Wigner-crystal type or bi-stripe type.

The diffraction peaks were searched in the direction along (h, h, 0) in the sample of x = 0.75. We could observe only the resonant scattering at q=(n±1/8, n±1/8, 0) and the resonant and nonresonat scattering at q=(n±1/4, n±1/4, 0). The signal from the lattice distortion of oxygen is not observed on this principle axis. We also observe the signal off this axis. These peaks come from the lattice distortion of oxygen. The same measurements in the sample of x = 0.67 have been performed. We have observed the resonant and nonresonant scattering at the modulation wave vectors δ = 1/6, 1/3 though the superlattice intensity is much weaker than that of x = 0.75. By comparing between the observed peak intensities and the model calculations, we have concluded that the charge is rather delocalized with a sinusoidal modulation and the orbital state has Wigner-crystal type in the low temperature phase of $Nd_{1-x}Sr_{1+x}MnO_4$ (x = 0.67, 0.75). The work has been done in collaboration with Y. Wakabayashi, H. Sawa, M. Kubota (PF, KEK) and T. Kimura, Y. Tokura (Tokyo Univ.).

Modulation of Orbital Ordering in $LaMnO_3$ Studied by Resonant X-ray Scattering

(T. Sato, H. Nakao, Y. Murakami)

The manganese ions of $LaMnO_3$ are all trivalent and have the orbital degree of freedom. The orbital of this compound is ordered at T_O=780 K and at the lower temperature T_N=140 K the compound becomes A-type antiferromagnetic. We have found that the peak position of the resonant reflection of (300) from the orbital order is shifted in the direction of H just below T_N. The shift was very small, about 10^{-3} of the reciprocal unit. The other resonant scattering peaks from the orbital order, (100) and (500) were measured. The same peak shifts were observed below T_N. This interesting result cannot be explained by a conventional incommensurate structure, because in the case we observe the peaks symmetrically around the fundamental Bragg peak. In this case we observe one peak asymmetrically. And also we cannot explain by a simple disordered model, because in the case we do not observe the peak shift and only the decrease of the peak intensity and the broadening of the peak width.

This phenomenon may be explained by an antiphase domain boundary (ADB) model of orbital states. However, it is not so easy to explain that the peak shift occurs only below T_N. One possibility is that the orbital disorder or x^2-y^2 state may be introduced because the A-type antiferromagnetic state favors the x^2-y^2 orbital state owing to the large transfer of electrons in the plane and no transfer inter-plane. If it should be true, the observed peak shift may be attributed to the competition between the orbital and the spin states. The work has been done in collaboration with Y. Wakabayashi (

PF, KEK), T. Arima (Tsukuba Univ.), Y. Tokura (Univ. Tokyo), K. Hirota (Univ. Tokyo), Y. Endoh (IMR, Tohoku Univ.).

Orbital correlation and excitation in $KCu_xZn_{1-x}F_3$

(S. Niioka, N. Tatami, H. Nakao, Y. Murakami)

The compound $KCuF_3$ is a typical orbital ordering system which is driven by Jahn-Teller effect. Our interest is how the orbital ordering disappears when the Cu ion is diluted by the non-magnetic ion Zn. We have observee the RXS intensities of $KCu_xZn_{1-x}F_3$ decreasing x from 1 to 0. We found that the signal disappears at x=0.6 which is very close to the two-dimensional percolation concentration. This disappearance of the orbital ordering is also confirmed by the structural phase transition from tetragonal to cubic at x=0.6. This concentration is curious because the system is three-dimensional. The orbital may work on the decrease of the dimensionality. We also observed the elementary excitation of $KCuF_3$ including the charge transfer excitation (2eV) by resonant inelastic x-ray scattering (RIXS) at SPring-8. The RIXS experiments of the diluted systems are planning to get the unique excitation near x=0.6 which comes from the fractal structure.

The work has been done in collaboration with Y. Wakabayashi, H. Sawa (PF, KEK), and K. Kuzushita, K. Ishii, T. Inami (JAERI).

Antiferro-quadrupolar ordering of 4f-electron state of $PrFe_4P_{12}$

(K. Iwasa)

$PrFe_4P_{12}$ is a novel Pr-based compound exhibiting a heavy-electron behavior. It undergoes a transition to non-magnetic ordered phase below 6.5 K, which is accompanied with a structural superlattice formation due to the Fe-ion displacement. Such low-temperature phase has been expected to be an antiferro-quadrupolar (AFQ) ordering of Pr-ion $4f^2$-electron state. In order to determine an order parameter, we performed neutron diffraction measurements under magnetic field parallel to the cubic crystal-axis [0, 0, 1] at JRR-3M reactor, JAERI. Tokai, Japan. By observing intensities of polarized neutron diffraction due to interference of scattering amplitudes from the Fe-ion displacement and the antiferromagnetic component induced along the applied magnetic field, we concluded that the dominant ordered quadrupole is $O_2^0 = (3J_z^2 - J(J+1))/2$ in the low-temperature phase. A future subject in the study of $PrFe_4P_{12}$ is to find the role of quadrupoles on the heavy-electron state. This work is in collaboration with Department of Physics, Tokyo Metropolitan University.

Characteristic magnetic excitation in the antiferro-quadrupolar ordering phase of $PrFe_4P_{12}$

(K. Iwasa)

Recently, the search for elementary excitations in the electron orbital ordered state has been a general and major subject in the field of strongly-correlated electron systems. As presented above, $PrFe_4P_{12}$ is a typical system showing the antiferro-quadrupolar (AFQ) ordering. Thus, we have investigated magnetic excitations in a single-crystal sample of $PrFe_4P_{12}$ by inelastic neutron scattering experiment. We observed sharp magnetic excitations in the AFQ ordered phase, whose maximum intensity appears at the Brillouin zone center and minimum at the zone boundary. Such collective magnetic excitation is expected to reflect an elementary excitation in the low-lying AFQ ordering. This work is in collaboration with Department of Physics, Tokyo Metropolitan University and Laboratoire Léon Brillouin, CEA/CNRS, France.

Drastic variation of the magnetic excitations on the metal-insulator phase transition of $PrRu_4P_{12}$

(K. Iwasa)

$PrRu_4P_{12}$ shows the metal-insulator transition at $T_{\mathrm{M-I}} = 63$ K accompanied with the upturn of electrical resistivity and the doubling of the crystal-structure periodicity. It is considered as a charge-density-wave formation, as suggested in the theoretical study of band structure indicating the three-dimensional Fermi-surface nesting condition. We performed inelastic neutron scattering experiment to investigate a role of 4f electrons in the M-I transition of $PrRu_4P_{12}$. Below about 20 K with less amount of carriers, there are resolution-limited sharp excitations represented by two kinds of crystal-field schemes of Pr ions due to the low-temperature superlattice formation. With increasing temperature, the excitation peaks shift by about 3 meV and broaden considerably up to 1.7 meV around $T_{\mathrm{M-I}}$. Such drastic evolution of the crystal-field excitations in $PrRu_4P_{12}$ indicates strong hybridization between 4f and conduction electrons. This work is in collaboration with Department of Physics, Tokyo Metropolitan University.

Magnetic-field-induced quadrupolar ordering in $PrOs_4Sb_{12}$

(K. Iwasa)

One of recent topics in the field of materials science is the so-called unconventional superconductivity mediated by magnetic or heavy-electron states. $PrOs_4Sb_{12}$ has been attractive because of the double superconducting phase transitions at 1.7 and 1.85 K and the proposed anisotropic gap suggested from the NQR and thermal conductivity under magnetic field. The unusually large jump of specific heat at the phase transition indicates the heavy quasiparticles to be responsible for the superconductivity. In addition, the other ordered phase under magnetic field above 4 T was pointed out by specific-heat measurement etc. By the neutron diffraction experiments in this field-induced phase, we observed antiferromagnetic component described as staggered magnetic moments normal to the magnetic field applied along the [0, 0, 1] axis. It is explained by the $O_{yz} = J_yJ_z + J_zJ_y$ antiferro-quadrupolar ordering with the crystal-field ground state Γ_1 and the first excited state $\Gamma_4^{(2)}$. Since such quadrupolar phase is located near by the superconducting phase, it is natural to expect that the quadrupolar fluctuation gives rise to the superconductivity. This work is in collaboration with Department of Physics, Tokyo Metropolitan University and CEA-Grenoble, France.

Multipole ordering in RB_2C_2 (R=Tb, Dy, Ho) studied by resonant x-ray scattering

(T. Matsumura, D. Okuyama, H. Nakao and Y. Murakami)

There are many magnetic materials in which the orbital degrees of freedom play important roles. We have been performing resonant x-ray scattering experiments to observe various behaviour of the quadrupolar moments of the $4f$-electron systems. Following the suceess of the observation of the antiferro quadrupolar ordering in DyB_2C_2, we have extended the study to HoB_2C_2 and TbB_2C_2 in which the competition between magnetic and quadrupolar interactions is more important for the physical properties. In TbB_2C_2, although the ground state is an antiferromagnetically ordered state, we have observed an ordering of quadrupole moments that accompany the antiferromagnetic order A challenge to observe more higher rank multipoles is going on in this system. The work has been done in collaboration with A. Tobo and H. Onodera (Department of Physics, Tohoku Univ.) and Y. Wakabayashi and H. Sawa (PF, KEK).

Relation between conduction electrons and crystalline electric field

(T. Matsumura and H. Ishida)

Strong correlation between conduction electrons and a crystalline electric field that localized $4f$ electrons experience has been studied experimentally in a system of $Tm_{0.05}La_xYb_{1-x}Te$. In $Tm_{0.05}La_{0.95}Te$ where the Tm ions are diluted in a metallic environment, large crystal field of about 100 K was observed. However, in $Tm_xYb_{1-x}Te$, an insulating system, the crystal field almost vanishs. This difference apparently comes from the existence of the conduction electrons, and we have been studying this relation by changing the number of conduction electrons with the La concentration x. With decreasing x the clear CF splitting decays into a broad excitation which reflects a distribution of the local environment of the conduction electron density. The experiments are performed using the LAM-D spectrometer at KEK with T. J. Sato (ISSP, Tokyo University).

Magnetic excitation of TmTe in the valence fluctuation state under high pressure

(T. Matsumura and H. Ishida)

TmTe is a divalent magnetic semiconductor at ambient pressure. Antiferroquadrupolar order and magnetic order take place at 1.8 K and 0.5 K, respectively. When the energy gap collapses under high pressure about 2 GPa, TmTe becomes metallic and, surprisingly, a ferromagnetism with T_c=15 K suddenly appears. We have tried to observe magnetic excitation spectrum in its metallic state by inelastic neutron scattering under high pressure. In the first experiment, the pressure was set just before the ferromagnetism appears. The result shows that the width of the quasi-elastic peak become broad, indicating an increase in single site spin fluctuation. The experiment was performed using the PONTA 3-axis spectrometer at JRR-3M with M. Nishi (ISSP, Tokyo University).

Measurement of Hall effect, magnetoresistivity, and magnetization

(T. Matsumura and H. Ishida)

To study transport properties of magnetic materials as a tool to understand magnetic and orbital correlations, we have developed a measurement system which is able to measure Hall resistivity and magnetoresistivity between 1.5 K and 300 K up to 10 Tesla. Two samples can be measured simultaneously. Temperature control, magnetic field sweep, and sample rotation are computer controled using LabView. Magnetization measurement system by extraction method is being developed.

Orbitally ordered state in $LaTiO_3$ studied by resonant x-ray scattering

(H. Nakao, S. Kodama, T. Sato, K. Kiyoto, D. Bizen and Y. Murakami)

Near Mott metal-insulator transition induced by the band-width and/or band-filling control, various physical properties, such as High-T_C superconductivity and colossal magnetoresistance, have been discovered and vigorously studied. The orbital degree of freedom as well as charge and spin often plays an important role in these phenomena. A perovskite-type transition metal oxide, $R\mathrm{TiO_3}$ (R: rare-earth element) has one $3d$-electron on Ti^{3+} ion and the electron has the orbital degree of freedom in the t_{2g} state. $\mathrm{YTiO_3}$ shows a typical orbital ordering of the t_{2g}-electron and the orbital ordering plays an important role in the ferromagnetic transition at $Tc \sim 30$ K. On the other hand, $\mathrm{LaTiO_3}$ shows G-type antiferromagnetic transition at $T_N \sim 150$ K. However, the orbital state is still a controversial problem. Therefore, we have investigated the orbitally ordered state of $\mathrm{LaTiO_3}$ utilizing the RXS near the Ti K-edge. On the basis of the RXS at $1s \rightarrow 3d$ transition energy (pre-edge), the orbital ordering is expected to be strongly suppressed as compared with the case of $\mathrm{YTiO_3}$. Moreover, we have also investigated the wave function of the ordered orbital and the change of the orbital state at the magnetic transition. This work has been done in collaboration with M. Kubota (Photon Factory, KEK), Y. Taguchi (Tohoku Univ.), and Y. Tokura (Univ. of Tokyo).

Charge distribution of Mn ion in artificial superlattices $[(\mathrm{LaMnO_3})_m(\mathrm{SrMnO_3})_m]_n$

(H. Nakao, J. Nishimura, S. Kodama, T. Sato, and Y. Murakami)

Fabrication technology of the artificial superlattice structure composed of different compositions has been rapidly developed and opens a new field of materials physics. In the transition metal perovskite system, artificially controlled charge ordered states have been also built up by the film deposition technique. Moreover, it is indicated that the peculiar physical properties of the superlattice are realized by the interface state because the valence state of the transition metal near the interfaces cannot be simply controlled by the stacking pattern. Therefore, the determination of the spatial charge distribution of the transition metal site becomes important for the study of the superlattices. In this study, we focus on the superlattices of $\mathrm{LaMnO_3}$ and $\mathrm{SrMnO_3}$, $[(\mathrm{LaMnO_3})_m(\mathrm{SrMnO_3})_m]_n$, where the manganese valence is expected to be varied from 3+ to 4+ and the physical properties depend strongly on the periodicity m To investigate the relation between the Mn valence state and the physical properties, we have tried to determine the charge distribution of the manganese ion using the anomalous x-ray scattering term of Mn ion near the K-edge. To verify the stacking structure of the superlattice, the x-ray diffraction along the stacking direction (c-axis) has been measured at E =6.52 keV (non-resonant energy). The satellite peaks originating from the superlattice periodicity and the sub-peaks due to the Laue function are clearly observed around the fundamental perovskite peak. The calculated diffraction pattern based on our stacking model shows good agreement with the experimental result. Moreover, we have estimated the charge distribution of Mn ion based on the energy dependence of the scattering intensity near Mn K-edge energy. This work is performed in collaboration with A. Ohtomo, T Fukumura M. Kawasaki (Tohoku Univ.), and T. Koida (ERATO).

Valence fluctuation state in the rare earth compound – Yb_4As_3 –

(H. Nakao, K. Kiyoto, S. Kodama, D. Bizen, T. Matsumura, K. Iwasa, Y. Murakami)

Valence fluctuation is one of the anomalous physical phenomena in the rare earth compound. Yb_4As_3 with anti-Th_3P_4 type cubic crystal structure shows typical valence fluctuated state. Only one kind of Yb ion crystallographically exists in the structure and the valence should be +2.25. However, the difference valence states, Yb^{2+} and Yb^{3+}, have clearly observed by x-ray absorption spectra, photoemission and reflectivity spectra. As a result, the thermally fluctuated valence state is expected in this compound. Moreover, the compound also shows the charge ordering of the Yb (2+/3+) ion at T_C ~290 K. Namely, it is expected that the fluctuating frequency decreases with decreasing temperature and the static charge ordered state finally emerges below T_C. However, the charge distribution in the valence fluctuated state has never been measured by x-ray diffraction technique. As a first step to study the valence fluctuated state, in this study, we have investigated the charge ordering of Yb ion using resonant x-ray scattering (RXS). In order to find the RXS owing to the charge ordering, the energy dependence of the scattering intensity at the several reflections was measured below T_C. We have clearly found the RXS at (3 -3 0) showing the resonant feature at the Yb L_3-edge. On the basis of the quantitative RXS intensity, we have tried to calculate the valence state at respective Yb site. This work is performed in collaboration with A. Ochiai (Tohoku Univ.).

Instrumentation

Project of installation of position-sensitive neutron detectors to TOPAN

(K. Iwasa)

Our laboratory takes care of the thermal-neutron triple-axis spectrometer TOPAN installed in the reactor JRR-3M in JAERI, Tokai, Japan, which is owned by Faculty of Science, Tohoku University. From July 2003, K. Iwasa has been a responsible person for this instrument. He has planned to install position-sensitive detectors covering wide scattering angle in the polarized neutron diffraction measurements. The PSD system developed in the pulsed neutron facility KENS in KEK, Japan will be combined with TOPAN near future.

Publications

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17. "Photoemission study of mixed-valent Tm-monochalcogenides: Evidence of electron-correlation effect in different Tm-core levels", K. G.

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20. "Studies of Unusual Magnetic and Electronic Properties of the Low-Carrier System CeP by Synchrotron X-rays", M. Kohgi, K. Iwasa, K. Kuwahara, A. Hannan, D. Kawana, Y. Noda, T. Shobu, K. Katsumata, Y. Narumi and Y. Tabata: Physica B **345** (2004) 55-58.

Master Theses (March 2003)

M1) Daisuke OKUYAMA: The Study on the competition between magnetic and quadrupolar orderings in TbB_2C_2.

M2) Soukichi KODAMA: Structural phase transition of $Y_{1-x}Ca_xTiO_3$ near the Metal-Insulator transition.

M3) Tadashi SATO: Charge and orbital modulation in $Nd_{1-x}Sr_{1+x}MnO_4$ and $LaMnO_3$.

M4) Shinya NIIOKA: Orbital correlation and excitation in $KCu_xZn_{1-x}F_3$.